ARTICLE

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Continuous decrease in soil organic matter despite increased plant productivity in an 80-years-old phosphorus-addition experiment

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The objective of this study was to understand how eight decades of tillage affected soil element dynamics. We measured soil chemical properties and the ^{14}C:^{12}C ratio ($\Delta^{14}C$) of organic carbon in one of the oldest cropland experiments in Europe with different levels of phosphorus addition. Soil total and organic phosphorus stocks in the uppermost 20 cm did not differ significantly between the control and the phosphorus addition treatments after 80 years, indicating plant phosphorus uptake from the subsoil. Crop yields increased from 220 g dry weight m^{-2} in 1936 to more than 500 g dry weight m^{-2} in the 2010s. The soil total organic carbon and total organic phosphorus stocks decreased by 13.7% and 11.6%, respectively, in the uppermost 20 cm of the soils during the experiment, irrespective of phosphorus addition. Based on modeling of $\Delta^{14}C$, we show that the mean transit time of carbon in the soil was below 10 years, indicating that a large share of the carbon inputs to soil is quickly respired. Our results suggest that the current agricultural practice at this long-term experiment is not sustainable because it led to a continuous decrease in soil organic matter over the last decades, despite increases in plant productivity.

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Long-term agricultural management must ensure that soil fertility is not compromised in order to meet sustainability goals. Most studies on sustainability of agriculture considered either soil element cycling or crop yields, but few studies have assessed the sustainability of agroecosystems over several decades combining these two aspects (with the exception of a few mostly modeling studies[1,2]). Further, there is only little observation-based knowledge about long-term effects of tillage on soil biogeochemistry and plant productivity since most experiments have only been conducted over periods of years up to a few decades. Therefore, the few existing long-term field experiments that have been maintained for more than half a century can provide very valuable insights into long-term effects of tillage, fertilizer application, and biomass removal on soil properties and yields, which is important as a basis for the development of sustainable agricultural practices[3,4].

In some long-term cropland experiments, it was found that soil total organic carbon (TOC) stocks decreased over decades, irrespective of application of organic or inorganic fertilizers; for instance, in Switzerland at the Zurich Organic Fertilization Experiment[5] or in the experiment at Woburn close to Rothamsted, UK[6]. The main reasons for these decreases seem to be the previous conversion of the sites to cropland that lead to changes in organic matter inputs in combination with destruction of aggregates caused by repeated plowing, which increases the decomposition rate[7-9]. Yet, in other experiments it was found that TOC stocks decreased only in the control treatment that did not receive nutrients, but increased in the treatments with inorganic nutrient application (and no organic matter addition); for instance in the Bad Lauchsted experiment, Germany[10,11]. These differences between control and fertilized plots are likely caused by differences in plant organic matter inputs to soil, resulting from differences in plant productivity.

The results regarding the effect of inorganic phosphorus (P) application on soil TOC stocks are rather ambivalent. P application often increases plant productivity, and thus can be expected to increase TOC stocks, particularly if added together with nitrogen, as in the Bad Lauchsted experiment[11] or the long-term experiment at La Estanzuela, Uruguay[12,13]. However, in some experiments, no significant effect of P fertilizer application on soil TOC stocks was observed, as in the Zurich Organic Fertilization Experiment[5]. Furthermore, a recent study of a long-term experiment at a grassland site reported that decades of fertilization with inorganic P increased soil organic matter (SOM) decomposition[14]. In addition, inorganic P fertilization under nitrogen limitation depleted soil TOC stocks in several Swedish long-term experiments[15]. The reason for this might be that addition of phosphate to the soil can cause desorption of adsorbed SOM, which is thereby rendered available for microbial decomposition[16,17].

Application of inorganic P might also affect the SOM stoichiometry and the size of different P pools in soil[18]. Specifically, decades of P application can change the concentration of plant-available P in the soil[19]. P application can also increase organic P stocks[18] and the content of specific organic P compounds, such as phytate, which is the most abundant organic P compound in soils[20,21].

The objective of this study was to understand how eight decades of agricultural production and P addition affected SOM stocks, SOM stoichiometry, soil P pools, and SOM decomposition in a temperate cropland. For this purpose, we studied an 80-year old long-term cropland experiment in the south of Sweden that comprises two P fertilization treatments and a control treatment and is one of the oldest agricultural experiments in Europe. We analyzed the development of the soil organic carbon, the stoichiometry of the SOM, several soil P pools, and the crop yields over time in three treatments (control and two P addition treatments). Further, we measured $\Delta^{14}C$ of soil TOC to elucidate the long-term dynamics of soil organic C. The analysis of $\Delta^{14}C$ of the TOC, in combination with modeling, is a powerful tool to understand TOC dynamics. This is particularly the case if samples are collected at different times from the same soil. The repeated sampling allows following how the so-called bomb ^{14}C, that is derived from nuclear bomb tests conducted in the 1960s, is incorporated into the SOM, as a tracer of modern carbon[14,22,23].

Results and Discussion

Soil organic carbon and plant productivity over time. Soil TOC, total nitrogen (TN), and total organic phosphorus (TOP) decreased linearly over five decades by 13.8–11.6% compared to their initial contents, irrespective of fertilization treatment (Fig. 1a, c, e, and Table 1). This decrease in the TOC stock of 0.14 t C ha^{-1} yr^{-1} observed here is in accordance with a long-term cropland experiment in Switzerland that started in 1949[5]. In this experiment, the TOC stock decreased by 0.10–0.25 t C ha^{-1} yr^{-1} in the uppermost 20 cm across all treatments, including those in which organic and inorganic fertilizers were added. The reason why the TOC stocks decreased in the experiment studied here is likely that plowing led to a disruption of aggregates, causing increased decomposition of SOM[7-9,24].

The parameter values obtained from a two-pool carbon (C) model with connection in series (Table 2) provide a reasonable representation of the temporal dynamics of the organic C stock and $\Delta^{14}C$, either with constant C inputs to soil or, in a second version of the model, with C inputs proportional to annual yields (Fig. 2b and c). The two models differ considerably in the size and decomposition rate of the two C fractions, indicating that the trends in C stocks and radiocarbon over time are likely due to a rearrangement of pool sizes and their decomposition rate, but with large uncertainty regarding the size and direction of this rearrangement. The model parameter $\gamma = 1.44$ indicates that the C input to the soil equals 144% of the C in the annual crop yields, a value similar to the one used in the second version of the model as a constant input (equaling 150% of the long-term average yield) based on data reported in Bolinder et al.[25]. The mean transit time of C in the soil, which quantifies the mean age of a C atom when leaving the soil, was below 10 years for both models despite differences in model parameters, and the median of the transit time distribution of C was below 7.5 years (Table 2). These transit time distributions indicate that new inputs entering the soil are respired relatively fast, with about 50% of the C being respired in a few years after entering the soil (2–6 years on average, Table 2), and only very small proportions staying for more than a decade.

There are only few estimates of C transit times for similar soils and ecosystems from previous studies. For a relatively similar agricultural topsoil (0–20 cm) in Sweden, Crow and Sierra[26], found a mean transit time of 21.9 years and a median transit time of 1.06 years, which agree well with our estimates. Using machine learning methods integrating different sources of ^{14}C data, Xiao et al.[27], reported mean transit times (age of respired C) between 100 and 500 years for northern Scandinavian regions, but this estimate considers the entire depth profile from 0 to 100 cm and may be biased by the presence of permafrost. Using output from Earth system models Sierra et al.[28], found that mean transit times are on average not larger than 67 years for surface soils, but with larger values for high-latitude regions. The mean transit time values that we obtained are relatively low for the latitudinal region, but are not very different from other estimates for transit times of C in agricultural soils[14,26].

The two versions of the model are not consistent in explaining the contribution of the fast and slow pools to the overall decline
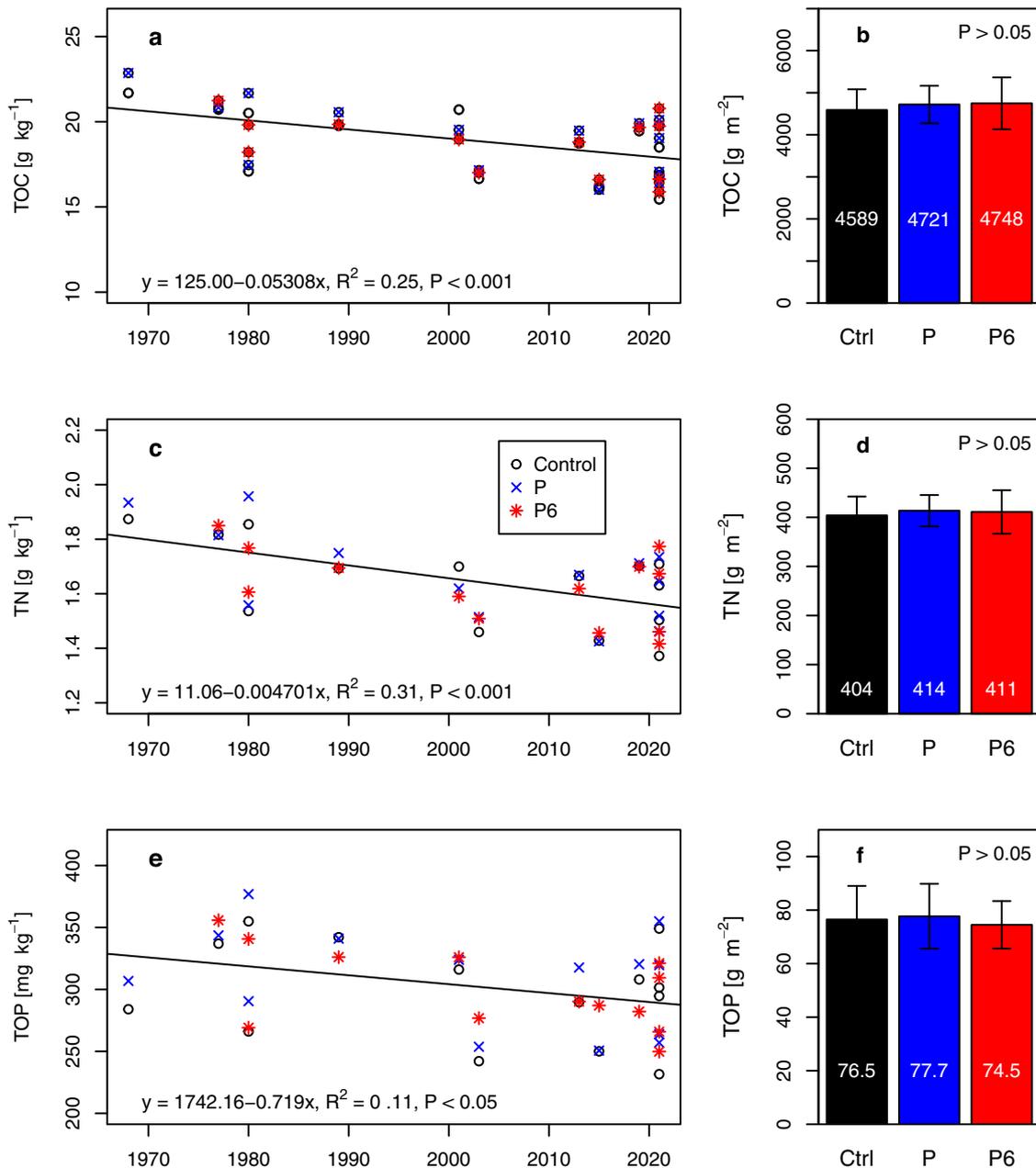
Fig. 1 Concentrations and stocks of total organic carbon (TOC), total nitrogen (TN), and total organic phosphorus (TOP). Concentrations of total organic carbon (TOC), total nitrogen (TN), and total organic phosphorus (TOP) as a function of time across the three treatments (**a, c, e**) and stocks of the same variables for the year 2021 separated by the three treatments (Control, P, and P6) of the field experiment (**b, d, f**). The concentrations as a function of time are approximated by linear models, for which the R^2 and the P value are given. The P values of the ANOVA of the stocks are given in the right corner of the right panels ($n = 4$). Error bars show standard deviations.

**Table 1 Stocks and losses of carbon, nitrogen, and different phosphorus pools.**

Variable	Stock 1968 (g m^{-2})	Stock 2021 (g m^{-2})	Loss from 1968 to 2021 (g m^{-2})	Loss per year (g m^{-2} y^{-1})	Decrease from 1968 to 2021 (%)
Total organic carbon (TOC)	5340.0	4608.6	731.4	13.80	13.7
Total nitrogen (TN)	470.2	405.4	64.8	1.22	13.8
Total organic phosphorus (TOP)	85.1	75.2	9.9	0.19	11.6
Total phosphorus (TP)	156.2	139.1	17.1	0.32	11.0
Phytate-phosphorus	26.7	23.0	3.7	0.07	13.8

Mean stocks of carbon, nitrogen, and different phosphorus pools in the uppermost 20 cm of the soil in 1968 and 2021 as well as the losses over 53 years (1968 – 2021), calculated across all three treatments (control, P, and P6) based on the linear models presented in Figs. 1 and 3.

Table 2 Parameter values obtained in the optimization for the two versions of the model.

Parameter	Model with constant input	Model with variable input
Decomposition rate of fast pool k_f (year^{-1})	0.115 ± 0.006	0.881 ± 0.108
Decomposition rate of slow pool k_s (year^{-1})	0.004 ± 0.003	0.004 ± 0.002
Proportional transfer from fast to slow pool α_{sf}	0.008 ± 0.008	0.004 ± 0.001
Proportion of annual yield as C inputs to soil γ	–	1.443 ± 0.48
Proportion of initial C in fast pool β	0.565 ± 0.051	0.094 ± 0.05
Mean transit time (uncertainty range) (years)	8.72 (7.04 – 9.87)	4.86 (1.07 – 11.64)
Median transit time (uncertainty range) (years)	5.86 (4.73 – 6.79)	3.29 (0.73 – 8.06)

In the version with constant input, the C input to soil was assumed to equal 150% of the long-term average yield. In the version with variable C input, the input varies with the yield from year to year, and the parameter γ determines the amount of C that enters the soil each year relative to the yield. Parameter values are reported as the mean ± standard deviation of posterior parameter values. Uncertainty ranges for mean and median transit times were obtained from maximum and minimum values of parameters' posterior distributions.
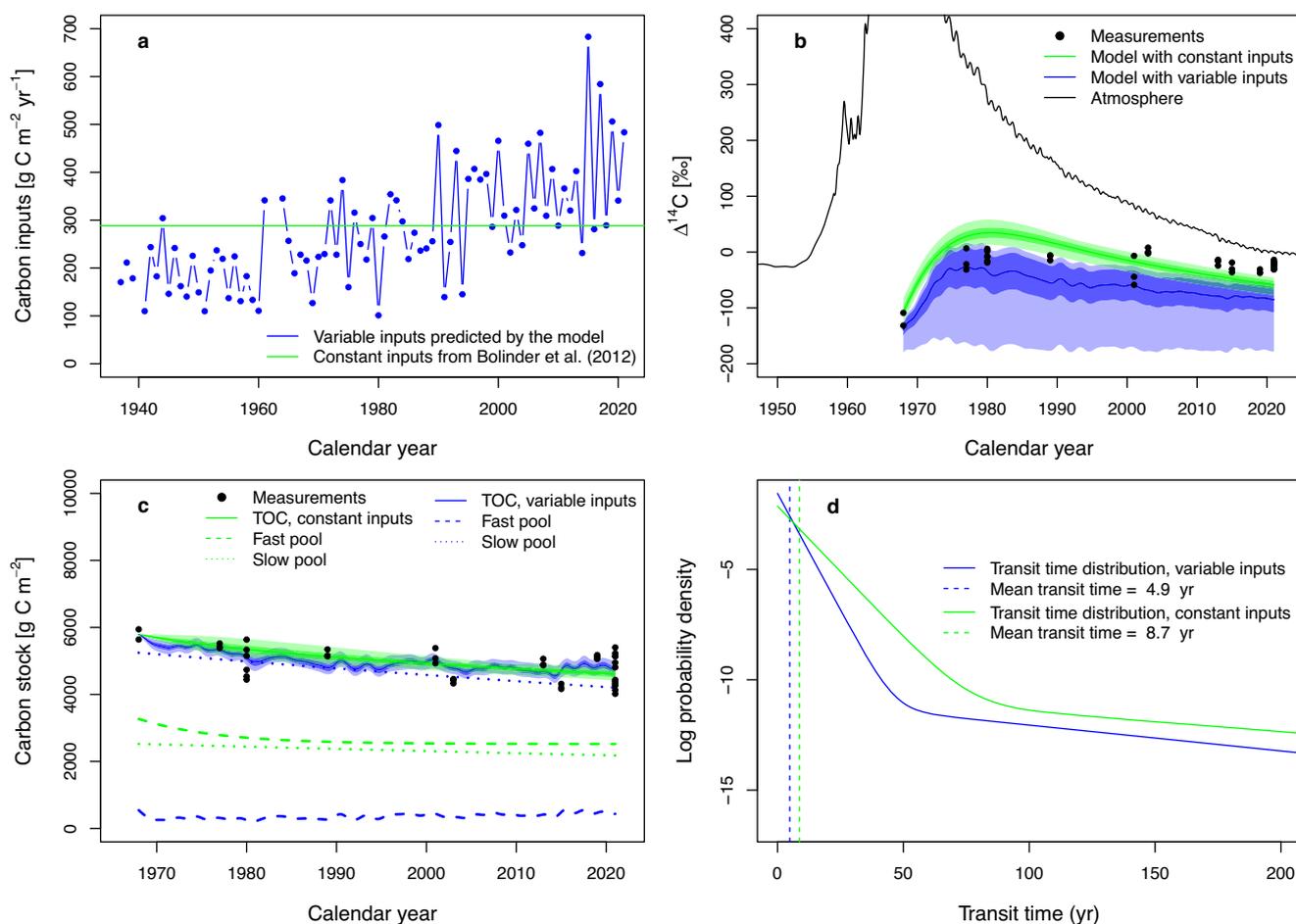
Fig. 2 Carbon inputs to soil, radiocarbon signature, carbon pools, and transit time of carbon in the soil. Carbon inputs to soil used in the simulations, either as constant input, computed as 150% of the average yield according to Bolinder et al. [25], or as variable input, computed as a constant proportion of the mean crop yields between 1936 and 2021, (**a**) together with radiocarbon signature of the soil total organic C (measured values and predictions by the two models) and the radiocarbon signature of the atmosphere (**b**), total organic carbon stocks of the soil and its two pools predicted by the two models (**c**) as well as the transit time distribution of organic carbon in the soil predicted by the two models (**d**). The data are derived from all three treatments of the field experiment (Control, P and P6). Uncertainty ranges of predictions in **b** and **c** were obtained by sampling the posterior distributions of parameter values according to their standard deviation (dark area) and the minimum and maximum values of the distribution (light area).

in TOC over time. While the model with constant inputs predicts an overall decline of the fast-cycling pool, the model with variable inputs predicts a significant decline of the slow-cycling pool. Regardless of whether modeled plant inputs are held constant or increase over time, the trends of TOC decline and bomb radiocarbon incorporation can only be explained by an increase in the decomposition rate of soil organic C. Our two-pool model represents this change as a shift in the relative contributions of the fast and slow-cycling pools to the total soil organic C pool. Although we observed an increase in plant productivity (yields) over the 80 years, it is uncertain if C inputs to soil have increased proportionally. The crop yield increased over time from about 220 g dry weight m^{-2} at the beginning of the experiment to more than 500 g dry weight m^{-2} in the last years of the experiment

(Fig. 2a). The increased crop productivity resulted from better pest control, more productive crop varieties, and potentially a slightly longer vegetation period over the last decades, as in many parts of Europe[29]. Our findings indicate that the soil organic C stock deteriorated despite an increase in plant productivity, which could have caused an increase in belowground C inputs to soil.

Our results are in agreement with other long-term experiments that observed decreases in soil TOC stocks in treatments despite increasing crop yields, such as the Zurich Organic Fertilization Experiment, Switzerland[5] and the long-term experiment at La Estanzuela, Uruguay[12,13]. At our study site, the decrease in SOM seems to have no effect on crop yields so far, likely because of the high nutrient contents in the subsoil (see below). However, if the SOM decline continues at a similar rate, it will likely affect soil fertility, and it might currently already affect other soil properties, such as water retention capacity and soil aggregation.

Soil organic carbon Δ^{14}C. The Δ^{14}C of the soil TOC in the beginning of the time series in 1968 was very low, -120‰ (Fig. 2b), indicating that the soil organic C was depleted in ^{14}C, hence old. The reason for this is likely the high clay content of the soil and the relatively cold climate, leading to low decomposition rates. Similarly Ellert and Janzen[30], reported that the Δ^{14}C of TOC was below -100‰ in the 1960s in the uppermost 15 cm of a cropland in the North American Great Plains with a climate similar to the one at our field experiment. In contrast, other studies on long-term observations of Δ^{14}C of soil TOC reported higher values even for the 1950s, before the bomb ^{14}C spike in the atmosphere, for sites in the UK and New Zealand[14,22,23]. This comparison suggests that the soil TOC at our site cycles slower than at other sites due to the colder climate.

We found evidence for the incorporation of bomb radiocarbon, even though the Δ^{14}C of TOC changed only slightly and stayed below 0 ‰ throughout the 53 years of observation, from 1968 to 2021 (Fig. 2b). Some previous studies in agricultural systems have reported larger values of Δ^{14}C incorporation after the 1960s[14,22,23]. Yet, our results are consistent with those in other ecosystems with low mean annual temperature and precipitation, where Δ^{14}C remained in negative values[30]. Low Δ^{14}C values after the 1960s, result from the fact that large stocks of old C require relatively large inputs of bomb ^{14}C in order to change substantially the isotopic signature of the TOC stock. Hence large Δ^{14}C values are more likely to be observed in soils with low organic C stock and large organic C inputs, as for instance in Stoner et al.[14]. Nevertheless, the trend of radiocarbon incorporation since the 1960s at our site was crucial to parameterize our soil C model, which helped to identify an overall increase in the turnover rate of organic C in the top 20 cm.

Soil phosphorus and crop yields. There was no significant difference in soil total phosphorus (TP) and TOP between the control and the P addition treatments in the uppermost 20 cm of the soils after 80 years of biomass removal ($P > 0.05$; Figs. 1f and 3b), which suggests that the crops (in the control treatment) took up P from the soil below 20 cm. The small and not significant difference in TP of 17 g m^{-2} between the control and the P treatments makes up only about 20% of the P that has been removed with the harvest over 80 years. Hence, substantial plant P uptake from the soil must occur below 20 cm. Plant P uptake from below 20 cm seems likely given that the soil P content increases strongly below 25 cm[31], which is related to the two different parent materials of the soil[31,32]. This finding calls into question the conclusion of a recent analysis on P fertilization trials, stating that the majority of P taken up by plants originates from the topsoil[33].

The P concentration of grains was on average 3.7 mg g^{-1} (Supplementary Fig. S1), and the yield, averaged over the whole period of the experiment, i.e., from 1936 − 2021, was 385 g dry weight m^{-2} yr^{-1}. Thus, 1.42 g P m^{-2} yr^{-1} were removed on average only due to the removal of yields (ignoring P removal due to harvest of leaves and stems). This value is below the fertilization rate of 1.75 g P m^{-2} yr^{-1} in the P addition treatment. Yet, the high yields beyond 600 g dry weight m^{-2} yr^{-1}, achieved during the last decade, particularly in the P fertilized treatment (Fig. 4) lead to a removal of more than 2.2 g P m^{-2} yr^{-1}, which is beyond the amount of annual P addition. The P required for the high yields of the last decade currently seem to be derived from the subsoil, both from natural P reserves and the surplus P that was added in the first decades of the experiment and likely accumulated in the very clayey subsoil.

P addition significantly increased the yields by 26 and 30% in the two P addition treatments throughout the period of the experiment, i.e., from 1936–2021 (Fig. 4). This increase in yields is very high compared to long-term P addition experiments in continental Europe. A recent meta-analysis found that the mean increase in crop yield did not exceed 4% in 30 P fertilization trials in Germany and Austria, for all levels of P fertilizer application[34]. The reason for the comparatively high responsiveness of the agroecosystem studied here to P fertilization is likely the high soil clay content which immobilizes a large part of the TP, and the fact that the soil has received less P fertilizer during the last century than many soils in continental Europe which were very strongly fertilized in the beginning of the second half of the 20th century[19].

Soil organic matter and phosphorus addition. We found that TOC stocks and Δ^{14}C did not differ significantly between the three fertilization treatments in 2021 ($P > 0.05$; Fig. 1b), indicating that increased plant productivity resulting from P fertilization (Fig. 3) had no significant effect on TOC stocks and organic C incorporation. The reason why we did not observe a decrease in TOC due to P application, as observed in other experiments[15,35,36], might be that the effect of phosphate addition on desorption of SOM plays only a subordinate role in soils in which SOM is largely stabilized by sorption to clay minerals and not to iron and aluminum oxides. This is supported by the fact that a negative effect of phosphate addition on organic matter sorption has been mostly observed in soils containing high concentrations of positively charged iron and aluminum oxides or allophanes, i.e., ferralsols, podzols, and andosols, but less in soils strongly dominated by clay minerals[17,37].

The P treatments differed significantly from the control treatment in plant-available P (P-AL), but not phytate-P and TOP contents in 2021 (Fig. 3b, d, f), indicating that a lack of P inputs in the control treatment does not lead to P mining of SOM, but enhanced plant P uptake from the P-AL pool. Decades of P addition had no significant effect on the SOM C:P ratio (i.e., the TOC:TOP ratio). The reason for this is likely that the TOC:TOP ratio was already very low in 1968 in the control, and thus further incorporation of organic P is rather unlikely. The molar TOC:TOP ratio was only 139 in the soil studied here, while on a global average, the topsoil of croplands in the temperate zone has a molar TOC:TOP ratio of 309[38]. The reason for the low TOC:TOP ratio in the soil studied here is likely the very fine texture (45% clay, see Table S1). This is supported by the observation that the clay-size fraction of the topsoil of croplands in the temperate zone usually has a much lower TOC:TOP ratio than the silt- or sand-size faction, on a global average of 197[38,39], which is relatively close to the TOC:TOP ratio of the bulk soil studied here. The clay likely stabilizes preferentially organic
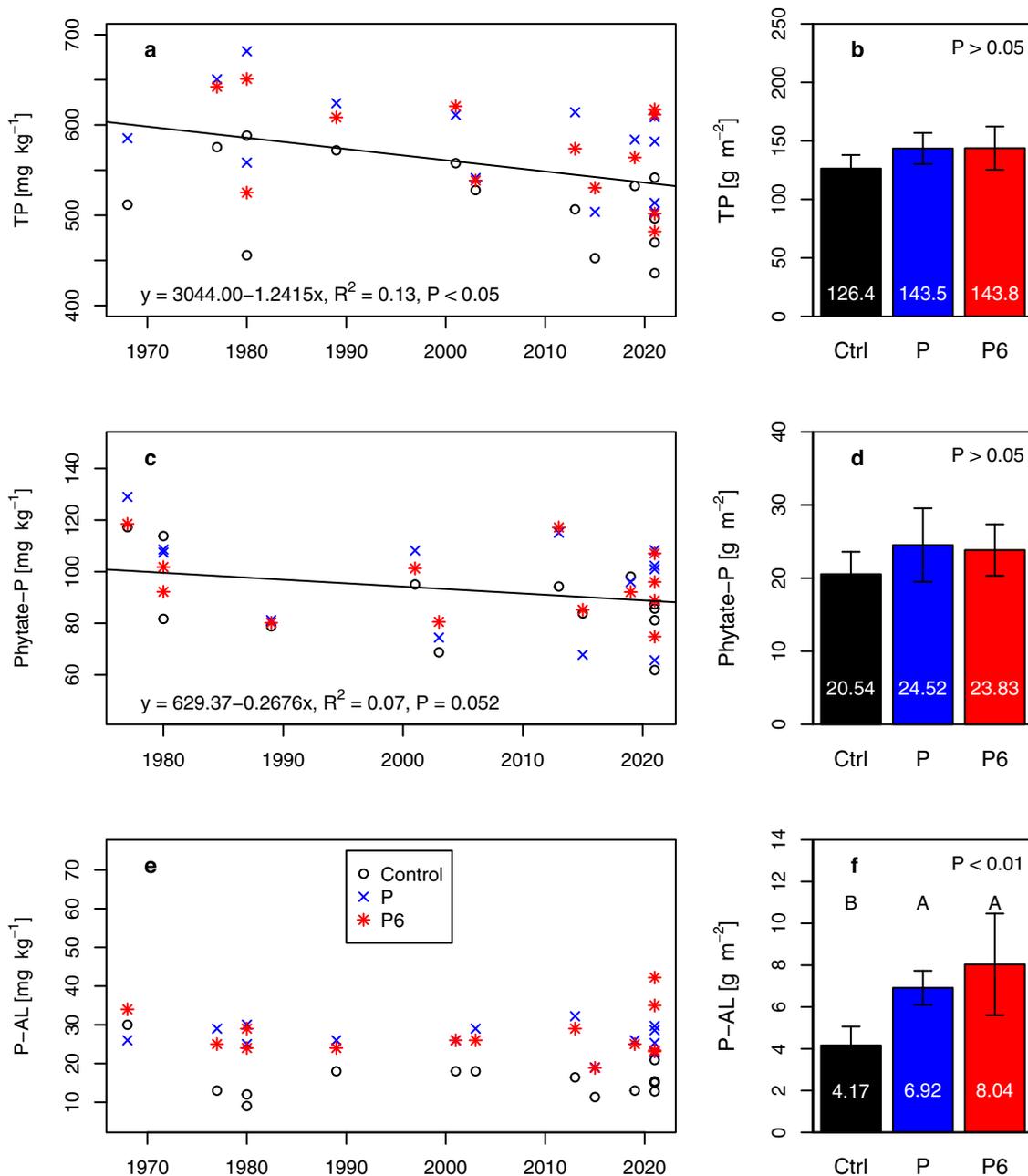
Fig. 3 Concentrations and stocks of total phosphorus (TP), phytate-phosphorus (phytate-P), and plant-available phosphorus (P-AL). Concentrations of total phosphorus (TP), phytate-phosphorus (phytate-P), and ammonium lactate extractable phosphorus (P-AL) as a function of time across the three treatments (**a**, **c**, **e**), and stocks of the same variables for the year 2021 separated by the three treatments (Control, P, and P6) of the field experiment (**b**, **d**, **f**). Linear models that describe the concentrations as a function of time are shown if $P \leq 0.055$, and the R^2 as well as the P value of the linear models are given in the panels on the left. The P values of the ANOVA of the stocks are given in the right corner of the right panels, and different letter indicate significant ($P < 0.05$) differences among the three treatments ($n = 4$). Error bars show standard deviations.

phosphorylated compounds because they have a high affinity to adsorb to soil minerals via their phosphate group, leading to a low TOC:TOP ratio in the clay fraction and in clayey soils[38-40]. Given that organic phosphorylated compounds adsorb strongly to mineral surfaces, which likely slows down their decomposition relative to non-phosphorylated compounds, we restrain from calculating organic P mineralization rates from our C model.

Further, only 31% of the TOP was formed by phytate-P. The reason for the relatively low contribution of phytate-P to the TOP pool in comparison to other soils[21] could be that the high clay content of the soil also stabilizes other phosphomonoesters and

phosphodiesters which might decompose substantially faster in soils with a more coarse texture.

Conclusion

We conclude that the high crop yields in the control treatment are based on plant P uptake from soil below 20 cm (and not from organic or inorganic P in the topsoil) since the soil TOP and TP stocks in the uppermost 20 cm in the control treatment do not significantly differ from those of the P addition treatments. We found that the soil TOC and TOP stocks decreased by 13.8% and 11.6%, respectively, in the uppermost 20 cm of the soils over the
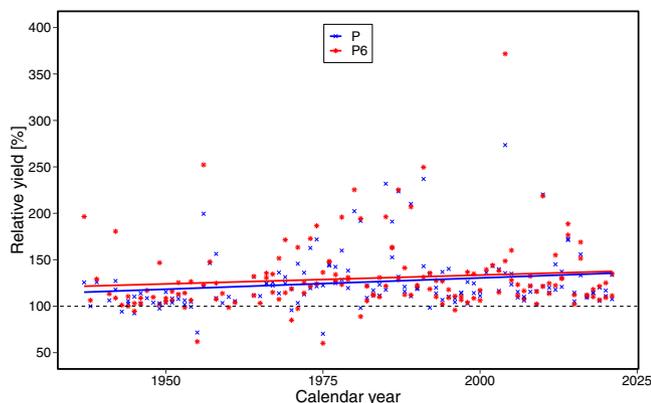
Fig. 4 Effect of phosphorus (P) fertilization on crop yield as a function of time. The relative crop yield (in %) in the two P addition treatments (P and P6) is calculated for each year as the yield in the treatment divided by the yield in the control multiplied by 100.

last 53 years, across all treatments. The results suggest that the agricultural use of the soil is not sustainable because it led to a continuous depletion of SOM that accumulated several decades to centuries before agricultural use of the soil. Based on modeling of $\Delta^{14}C$, we show that the mean transit time of C in the soil was below 10 years, indicating that a large share of the C inputs to soil is quickly respired, while only a small proportion of the C input remains in the soil for several decades. This finding suggests that the soil TOC content only responds very slowly to changes in the organic C inputs to soil. Our study demonstrates that long-term (>50 years) agricultural experiments are crucial to understand element cycling in the plant-soil system over time-scales relevant for sustainable development of agroecosystems.

Material and methods
Field experiment. The long-term field experiment is located in South-West Sweden (58°20'49.9"N and 13°07'36.1"E) at the field station Lanna, which is maintained by the Swedish University of Agricultural Sciences (SLU). Mean annual temperature and mean annual precipitation were 6.1 °C and 558 mm in the period 1961–1990, and 7.3 °C and 584 mm in the period 1991–2020. The experiment is located at 75 m a.s.l. on a plain of Quaternary glacial clay deposits. The glacial clay is overlain by an approximately 30 cm thick postglacial silty clay loam[32]. The area was drained in the late 19th century to enable crop production[31]. The soil is classified as an Udertic Haploboroll with a silty clay topsoil and a clay subsoil both with strong coarse subangular blocky structure[41]. The soil in the uppermost 20 cm is very fine-textured, and contains 42% clay and 15% fine silt (Table S1). The soil pH was on average 6.3 over the 53 years of observation, and there was no significant effect of P application (see Supplement Fig. S2).

The experiment consists of two identical halves, one started in 1936 and the other in 1941. In each of the two halves of the experiment, each treatment is replicated twice (total $n = 4$). (Each treatment used to be replicated four times in each half of the experiment until the 1970s when two replicates per experimental half were used for a new, additional lime treatment.). For the present study, only three treatments were considered. A control treatment that does not receive P, a P addition treatment (called P treatment in the following) in which 1.75 g P m^{-2} yr^{-1} is applied, and a second P addition treatment (called P6 in the following), in which the same amount of P per year is added but concentrated every six years, i.e., 10.5 g P m^{-2} every 6th year. P is added as superphosphate. In addition, all three treatments (Control, P, and P6) receive Ca(NO$_3$)$_2$ annually. The plot size is 7.0 m × 7.0 m.

The experiment (which has the reference number R3-1001) comprises also lime treatments that are not considered here, but were recently analyzed regarding the effects of liming on soil P dynamics by Simonson et al. (2018)[42] and effects of liming on crop yields by Börjesson and Kirchmann[43]. During the first 20 years of the experiment, a fixed seven-year crop rotation was maintained, consisting of winter wheat, peas, spring cereals, ley, ley (2nd year), spring cereals, and fallow. During each rotation, all plots were fertilized twice with farmyard manure at a rate of 2.0 kg m^{-2}. Since the late 1950s, the rotation has changed, now consisting mainly of cereals and occasionally oilseed rape and fallow. Manure is no longer used since the 1950es and no P fertilizer other than the experimental treatment is applied. The soil is plowed once per year, down to 20 cm, and crop residues are left on the field.

Data and sample collection. Crop yields have been measured (almost) every year since the beginning of the experiment. Soil samples from the uppermost 20 cm have been collected every six years before P fertilization. These samples were air-dried, and sieved (<2 mm) whereby roots were removed from the soil. Until 2019, soil samples of the replicates of each treatment were combined prior to the chemical analyses (pH and P-AL) and archiving of the samples. Archived soil samples from all three treatments (Control, P, and P6) exist from the years 1968, 1977, 1980, 1989, 2001, 2003, 2013, 2015, and 2019, with the exception that from 1968 only two samples (Control and P) have been conserved. In addition, the total P content of the yields (grains) was determined every year for the period 1995–2009.

For the present study, soil samples from all four replicates of the three treatments (Control, P, and P6) were collected. Specifically, one composite sample consisting of five cores of the uppermost 20 cm of each plot was taken in the autumn of 2021. These samples were air-dried, and sieved (<2 mm) whereby roots were removed from the soil. In contrast to the samples of the previous years, samples of all replicates were analyzed separately. Together with the archived soil samples, the soil samples examined in the present study cover a period of 53 years (1968–2021) and form a 10-point time series.

Chemical and isotope analyses. Plant available P (P-AL) was extracted from the soils by acid ammonium lactate according to the method of Egnér et al.[44]. Briefly, 5 g soil were shaken for 1.5 h in a solution containing 0.1 M ammonium lactate and 0.4 M acetic acid, with a pH adjusted to 3.75. The suspension was filtered (0.2 μm) before analysis using inductively coupled plasma-optical emissions spectroscopy (ICP-OES; Avio 200, Perkin Elmer).

The pH was measured after suspending 6.0 g soil in 18 ml H$_2$O.

Organic P was determined according to Saunders and Williams[45] and Williams and Saunders[46] as specified in Pansu and Gautheyrou[47]. Briefly, each sample was separated in two aliquots, each of 1 g. The first aliquot was directly extracted in 0.5 M H$_2$SO$_4$ on a horizontal shaker for 16 h. The other aliquot was ignited at 550 °C for 2 h and subsequently extracted in H$_2$SO$_4$ in the same way as the non-ignited sample. Inorganic P was measured in the extracts by the molybdenum blue method according to Murphy and Riley[48] using a continuous flow system (AA500, Seal). Total organic P was calculated as the difference between inorganic P in ignited and non-ignited samples. P in the non-ignited sample is considered as total P (TP).

Inositol-hexakisphosphate (called phytate in the following) was determined in the soil according to Turner[49]. Briefly, phytate was extracted from the soils in a solution containing 0.25 M NaOH and 50 mM EDTA. It was isolated from other organic phosphates

by hypobromite oxidation. Total P in the solution of the purified phytates was determined using ICP-OES (Avio 200, Perkin Elmer).

The total organic C and total N were measured by combustion using a LECO CNS-2000 analyzer. In addition, also inorganic C was measured using a LECO CNS-2000 analyzer in order to confirm that the soils were free of carbonates.

The $^{14}C:^{12}C$ ratio of TOC was analyzed using the accelerator mass spectrometer MICADAS (Ionplus, Dietikon, Switzerland) at the Max Planck Institute for Biogeochemistry in Jena, Germany. The radiocarbon ratio is reported as $\Delta^{14}C$ in per mille [‰], which is the fraction with respect to the standard isotope ratio (oxalic acid standard SRM-4990C; Steinhof et al., 2017[50]) including the normalization for $\delta^{13}C$ (fractionation correction) and the correction for the decay between 1950 and the measurement time[51].

For the texture analysis, samples from all three treatments of each half of the experiment were combined. They were pre-treated with H_2O_2, and then the particle size distribution was determined by wet sieving and sedimentation.

The P content of the yield was assessed by boiling the milled grains in 65% HNO^3, and the filtered digest was analyzed for total P using ICP-OES (Avio 200, Perkin Elmer).

Calculations and statistics. Stocks were calculated for the uppermost 20 cm of the soils based on a bulk density of 1.3 g cm^{-3} reported by Simonson et al. (2018)[42]. Thus, our calculation of stocks does not capture potential changes in bulk density that might have occurred during the field experiment. The relative yield (in %) of the two P addition treatments was calculated for each year as the yield in the treatment divided by the yield in the control multiplied by 100. We tested for significant differences in the soil chemical properties among the three treatments in 2021 by ANOVA followed by Tukey's test, and we considered $P < 0.05$ as the threshold for significance. The residuals of the ANOVA model were tested for normality using the Shapiro–Wilk test and for variance homogeneity using Levene's test in order to verify that the assumptions of normality and variance homogeneity were met. We calculated linear and monoexponential models for the soil chemical properties and relative yield as a function of time. The fit of the linear model and the exponential model that describe the soil element contents as a function of time were compared using Akaike Information Criterion (AIC) and the residual standard error. Both models fitted similarly well for all soil variables (Table S2). All data analyses were conducted using R (version 4.1.1, R Core Team, 2021[52]).

Modeling. We used data on soil TOC stocks, radiocarbon in bulk soil, and annual yields from all three treatments to inform a two-pool model that represents the dynamics of a fast and a slow C pool (C_f and C_s). The model can be expressed as

$$\frac{dC_f}{dt} = \gamma Y - k_f C_f$$
$$\frac{dC_s}{dt} = \alpha_{sf} k_f C_f - k_s C_s$$

with initial conditions $C_{f0}(t=0) = \beta C_0, C_{s0}(t=0) = (1-\beta)C_0$; where C_0 is the initial TOC content, and β is the proportion of the initial TOC in the fast pool. Y represents the annual yield (in units of g Cm^{-2}), γ is a proportion, and γY the amount of yield that equals the C input to the soil. The decomposition rates for the fast and slow pools are represented by the symbols k_f and k_s, respectively. The model assumes that a proportion of the organic carbon is transferred from the fast pool to the slow pool, which is expressed by the symbol α_{sf}, and is often called a 'humification'

or 'carbon use efficiency' coefficient. The model is similar in structure to the ICBM model[3] which has been used extensively to represent C and N dynamics in Swedish agricultural soils.

Note that in the model presented above, C inputs to soil are proportional to the annual yield. However, it is possible that the C inputs to soil did not increase linearly with the crop yields. Therefore, we tested additionally a second version of the model in which C inputs are constant over time. Following Bolinder et al.[25], who reported data on relative C allocation to yield and to total C inputs to soil, we calculated the C input as 150% of the C allocation to yield (and the yield was calculated as the average yield over the total duration of the experiment). Thus, the two versions of the model represent two contrasting assumptions, either constant C inputs to soil (despite increasing yields) or linear increases in C inputs along with increases in yields.

We used the R package SoilR[53] to implement a radiocarbon version of the models, and used the R package FME[54] to obtain parameter values using inverse data-assimilation in two steps; with a classical optimization in a first step to obtain priors for parameter values, and with a Markov chain Monte Carlo procedure to obtain posterior parameter values in a second step. We used the TOC stock and radiocarbon data in the cost function and obtained sets of best possible values for γ, k_f, k_s, α_{sf}, and β with their respective uncertainty. The model was driven by the time series of annual yields and the atmospheric bomb curve[55]. The optimization procedure searched for the combination of parameters that minimize the error between model predictions and observations of TOC stocks and soil radiocarbon. To obtain the transit time distribution predicted by the model, we used the best set of parameters of the model and applied the equations for transit time distributions described in Metzler & Sierra[56].

We refrain from presenting radiocarbon ages in years because in open systems, such as soils, radiocarbon is continuously exchanged with the atmosphere and traditional radiocarbon dating cannot be done[57].

Data availability
All data are available at this repository. https://doi.org/10.5281/zenodo.8070747.

Code availability
All code for reproducing the results is available at this repository. https://doi.org/10.5281/zenodo.8070747.

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Author contributions
M.S. and C.A.S. conceptualized the study, analyzed data, and wrote the manuscript. S.B. leads the long-term field experiment and contributed to data analysis and writing.

Competing interests
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